Structure and Thermodynamics of Curved Interfaces.

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In this paper we present new analytical results for the structure and thermodynamics of curved interfaces. Starting from a density functional formalism, we show that the square gradient approximation is fully consistent with the capillary approximation for the thermodynamics of droplet formation, namely i) a meaningful droplet radius may be defined whose size is dictated by the Laplace pressure difference across the interface, ii) provided the droplet is large, such pressure difference is accurately obtained from bulk equation of state data, iii) the work of formation is one third of the droplet's surface free energy. Using an approximate model which is asymptotically exact, we show that two different regimes exist depending on the droplet's size relative to the interfacial width. For large droplets, corrections to the classical capillary approximation are exponentially small, and an asymptotically exact expansion for the Tolman length in powers of the inverse droplet radius may be obtained. For droplets with size of the order of bulk correlation lengths, expressions for the Tolman length are still possible, but they no longer adopt a simple power law.